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Bistable resistive memory behavior in gelatin-CdTe quantum dot composite film

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Abstract. Bistable memory behavior has been observed for the first time in gelatin type A thin film dispersed with functionalized CdTe quantum dots. The two terminal device with the polymer nanocomposite layer sandwiched between an indium tin oxide coated glass plate and an aluminium top electrode performs as a bistable resistive random access memory module. Butterfly shaped (O-shaped with a hysteresis in forward and reverse sweeps) current-voltage response is observed in this device. The conduction mechanism leading to the bistable electrical switching has been deduced to be a combination of ohmic and electron hopping.

Keywords. Electrical bistability, biodegradable, conduction mechanism, resistive random access memory.

INTRODUCTION

Resistive random access memory (ReRAM) is an emerging nonvolatile memory which has drawn the attention of researcher mainly due to its simple design and competitive performance[1]. ReRAM architecture with an active resistive layer sandwiched between two electrodes is popular due to its simplicity and a major part of the research done so far has been on inorganic active layers. ReRAM modules with polymer nanocomposite active layer with excellent bistability, high storage density, high endurance and large retention time have been recently demonstrated[2-8]. In search for a solution to tackle the astronomically accumulating electronic waste, scientists have come up with new organic and biomaterials with excellent biodegradability for electronic device applications[9-18]. Gelatin, a polymeric biomaterial substance made of protein with aminoacids has shown great potential for ReRAM applications[17,18]. Electro chemical analyses of Type A Gelatin have been extensively carried to establish its electronic applications[19]. The heteroatom chemical structure of gelatin containing nitrogen reacts with metal ions to induce flow of electrons when used as active layer of ReRAM modules[20,21]. Dispersion of appropriate nanomaterials in polymers and biomaterials has resulted in high performance memory behavior[18,22]. Among the plethora of nanomaterials, semiconducting CdTe quantum dots have been used as dispersion in an organic matrix to increase the electrical conductivity has resulted in bistable memory device[23,24]. Since gelatin has not been well explored for ReRAM applications despite its low cost, abundance and good biodegradability, we were motived to explore its potential for the application in pure form and also as a composite wth nanocrystalline CdTe as the dispersant.

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MATERIALS AND METHODS

Gelatin of type A with Bloom strength of 300 and purity of 99.5% was purchased from Goodrich ingredients and chemicals Pvt Ltd, Chennai, India. CdTe core quantum dots functionalized with carbozyl group was purchased from Sigma Aldrich and used as the dispersant. The functionalized quantum dots are hydrophilic and easily disperses in aqueous solution. 5 w/v% aqueous solution of gelatin A was first prepared. 0.5wt/v % CdTe was dispersed in in the gelatin solution and the composite mixture was ultrasonicated for 15 min. to ensure uniform dispersion. A few drops of the homogenized solution was dropped on a pre-cleaned indium tin oxide (ITO) coated glass plate. The drops were spin coated in two steps viz., 500 rpm for 15 sec and 2500 rpm for 30 sec so that a uniform layer of the composite was formed over the ITO. The coated glass plate was then dried at room temperature. A home-made thermal evaporator was used to deposit the top aluminium metal electrode of area 0.5 cm2 under a vacuum of <10-5 mbar. Electrical measurements were carried out using a semiconductor parameter analyzer (Keithley 4200-SCS). In a typical experiment, the current-volatage (I-V) characterisicts of the sandwich structure was measured by scanning the voltage from -8 V to +8 V (forward sweep) followed by a reverse sweep (+8 V to -8 V). A scanning electron microscope (SEM, Jeol, JSM 6360A) was employed to examine the surface morphology of the thin film layers and to confirm the homogeneity of the films. Thickness of the films was measured using a thin film analyser (Filmetrics F10). thin film analyser. Both the pure gelatin A and the composite active layers were ~600 nm since the solutions had similar viscosity and the drops were spin coated under similar conditions.

RESULTS AND DISCUSSION

Fig. 1 shows the semi-logarithmic plot of I-V characteristics of the device with the prestine Gelatin A film as active layer. It is obvious from the flat response in figure that this device does not show resistive switching. Interestingly, resistive switching behaviour has been reported[17,18] in similar device structure with gelatin B as active layer. Hence, it appears that all types of gelatin may not show the resistive switching and that this phenomenon may be limited to only to gelatin B. However, gelatin A with 0.5 w/v% CdTe shows a different I-V characteristics as depicted in Fig. 2. The arrows in the figure show the sweep direction. Fig. 2 shows O-shaped I-V response[25] but with hysteresis behaviour (butterfly shaped). Similar hysteresis in O-shaped I-V response was earlier reported in the Pt/MoO_x/ITO system by Wang et al.[26] In this type of resistive switching behaviour, no abrupt increase of current is seen at a specific voltage. Instead, the write voltage either increases or decreases the conductivity.This is also different from the S-shaped behaviour reported in the prestine gelatin B[17,18] device, in which an abrupt increase or jump in current at a particular voltage was observed.

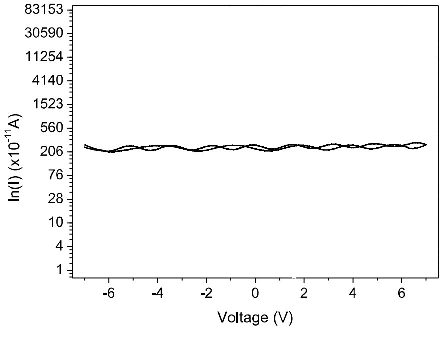

FIGURE 1: I-V characteristics of device with pure gelatin A film as active layer.

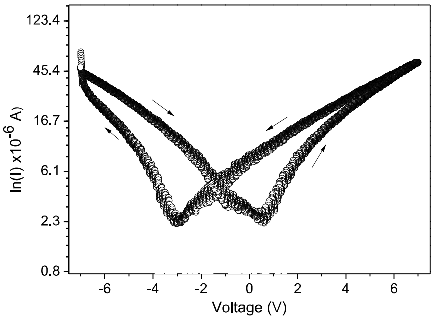

FIGURE 2. I-V characteristics of device with gelatin A and CdTe composie film as active layer.

It is mandatory to understand the conduction mechanisms involved in the resistive switching memory systems before assessing their potential for commercialization[27]. In most cases the current follows the relation, ln(I) $\propto V^{1/2}$ in the low voltage regime corresponding to thermionic emission and at higher voltages, ln (I) ~ ln(V) with a slope ~ 2. The latter refers to space charge limited conduction mechanism. Trap controlled space charge limited conduction can also occur in some cases when In(I) $\propto V^{\alpha}$, where $\alpha > 2$. We analysed the I-V data of the composite film device in terms of different mechanisms such as ohmic, space charge limited conduction etc. The low voltage data could be fitted well to a linear ohmic behaviour as shown in Fig. 3 and the high voltage data showed an excellent fit an exponential function of the type $I = I_o + A\exp\{(V-V_a)/KT\}$, indicating hopping type of conductivity[28] as depicted in Fig. 4. Though Fig. 3 and 4 present the analyses done on I-V data recorded in the forward sweep, the reverse sweep also showed the same behaviour (not shown here). Thus, the conduction mechanism in this system initially follows the linear hohmic behaviour due to conductive path provided by the sparcely distributed CdTe quantum dots at low voltages. Then, formation of conduction filaments and the rupture of the same can lead to low resistance and high resistance states. Such a situation can occur when electron hops from the electrode to a quantum dot or from one quantum dot to another and then from a quantum dot to the other electrode, essentially through a hopping mechanism. The conduction mechanism in Gelatin-CdTe films is different from the other systems that show O-shaped memory behaviour such as PMMA:ZnO[29] and Ag-PMMA[30] systems. It should also be noted that the earlier reports on the memory behaviour in CdTe system is with reference to capacitive memory and not resistive memory effect[23]. Further, the charge storage on quantum dots was attributed to the bistable memory behavior[23,24,31,32]. This comparison highlights the fact that all resistive memory devices exhibiting O-shaped memory behaviour may not have the same conduction mechanism.

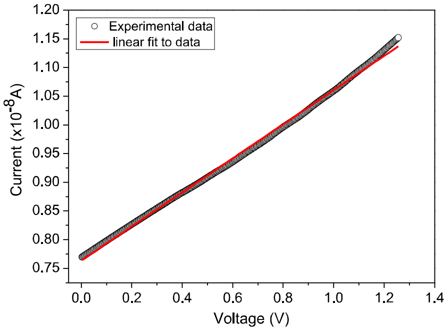

FIGURE 3. Linear (ohmic) fit to the low voltage regime (forward sweep) of I-V data of composite film.

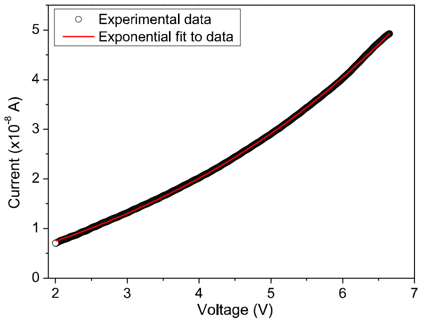

FIGURE 4: Exponential fit to the high voltage regime (forward sweep) of I-V data of composite film.

CONCLUSION

ReRAM modules have been fabricated and tested with prestine gelatin A and composite (gelatin A + CdTe quantum dots) films of ~ 600 nm thickness as active resistive layer. While the device with prestine gelatin A layer did not show resistive switching, the device with th composite film exhibited O-shapted resistive switching behaviour with hysteresis. This is the first report of resistive switching behaviour in this system. Analysis of the I-V data of the ReRAM module reveals a two step conduction mechamism leading to the resistive switching characteristics. Optimization of the device by varying the concentration of CdTe and the active layer thickness may help in realizing high performance ReRAM modules in this system.

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